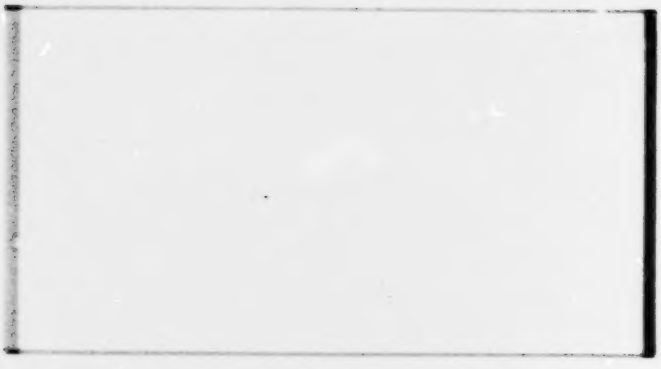
Department of Metallurgy & Materials Science



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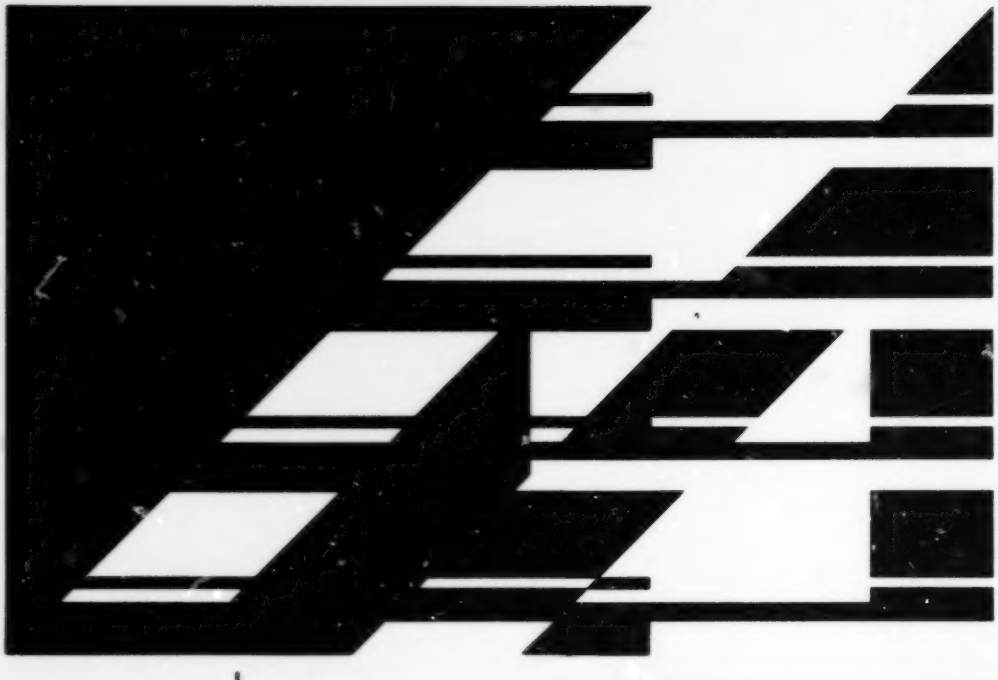

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SURFACE TENSION AND DENSITY OF THE MOLTEN $PbCl_2$-KCl-NaCl TERNARY SYSTEM

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ABSTRACT

The maximum bubble pressure method was used to determine the surface tension and density of melts within the $PbCl_2$-KCl-NaCl system. The temperature range of this study was from 450 to 800 ^{o}C. In all cases, the surface tension was found to decrease with increasing temperature. At constant molar ratio of KCl to NaCl, a minimum in the surface tension was observed at approximately 40 mole percent $PbCl_2$. The ternary surface tension values were found to obey the simple additivity expression of the binary surface tensions of $PbCl_2$-KCl and $PbCl_2$-NaCl. Based on these findings, constant surface tension contours have been drawn. In a similar manner the surface tension in the ternary $PbCl_2$-KCl-LiCl has been estimated.

The density obtained in the present study agree well with the previously determined densities using a bottom-balance Archimedean technique reported by this laboratory.

INTRODUCTION

The present process for the extraction of lead from lead sulphide concentrates involves sintering followed by high temperature carbon reduction in a blast furnace. Although this process is effective for lead production from high grade lead concentrates, it does give rise to problems with respect to environmental concerns and to the treatment of complex lead ores. The public concern of the environmental effects of lead extraction will no doubt introduce strict legislation on the lead content in fumes and surroundings, as well as sulphur dioxide emissions.

In anticipation of such regulations, new processes have been proposed. One such process is the conversion of lead sulphide to a chloride followed by fused-salt electrolysis of the lead chloride to produce metallic lead and chlorine(1-3). The chlorine produced is recycled directly or indirectly for the production of lead chloride. Various steps in this process are being actively investigated by CANMET.

Fused-salt electrolysis for the recovery of lead poses a number of problems. Lead chloride alone is not a good electrolyte. It has a high vapour pressure and exhibits poor current efficiency with respect to lead production. However, the addition of alkali chlorides decreases the fuming of lead chloride and improves the electrical conductivities. The favoured electrolyte composition appears to be the $PbCl_2$-KCl-LiCl ternary

system. One problem with this system is the hygroscopic nature of LiCl. For this reason, the $PbCl_2$-KCl-NaCl ternary system has been suggested and serves as an alternative electrolyte inspite of its slightly lower molar conductivity.

Unfortunately, the physical properties of these ternary system are scarce. Recently, we have determined the densities and molar volumes of the melt in both the $PbCl_2$-KCl-NaCl and $PbCl_2$ -KCl-LiCl ternary systems through a DSS contract with CANMET(4). During this study we noted that alkali chloride melts containing PbCl tended to creep up the crucible wall. Such observations suggest that the surface tension of these melts is quite low and could give rise to creeping of the electrolyte during cell operation. This phenomenon could lead to poor current efficiency. However, surface tension data on these systems are not available in the literature.

Consequently, the present study was undertaken to determine the surface tension of the molten salt system $PbCl_2$-KCl-NaCl as a function of composition and temperature in the range of interest for the electrolytic process of lead recovery.

EXPERIMENTAL

Several techniques have been considered with regard to the suitability for the determination of the surface tension of the molten salts.These include the maximum bubble pressure, Wilhelmy slide plate, capillary rise, maximum pull on cylinder, pin method, pendant drop method, ring method and the sessile drop technique. The numerous problems associated with interfacial energy measurements over the temperature range of interest have lead to the choice of the maximum bubble technique. One of the advantages of this method is that the density of the liquid salt can be measured simultaneously with the surface tension. This method is also capable of giving accurate and reproducible values at high temperatures in a moderately corrosive liquid, and has been used in many investigations(5,6,7) of the surface tension of molten salts. Furthermore, the principles of this method are well established.

It involves the slow formation of a gas bubble at the tip of a capillary immersed in the liquid. Simultaneously, the pressure of the gas used to form the bubble is measured in order to determine the maximum pressure developed inside the bubble at the very instant it bursts. Schrodinger(8) developed the following relationship between the surface tension and the maximum bubble pressure.

$$\gamma = \frac{rP_\gamma}{2}\left[1 - 2/3\left(\frac{r\rho g}{P_\gamma}\right) - 1/6\left(\frac{r\rho g}{P_\gamma}\right)^2 \ldots\right]$$

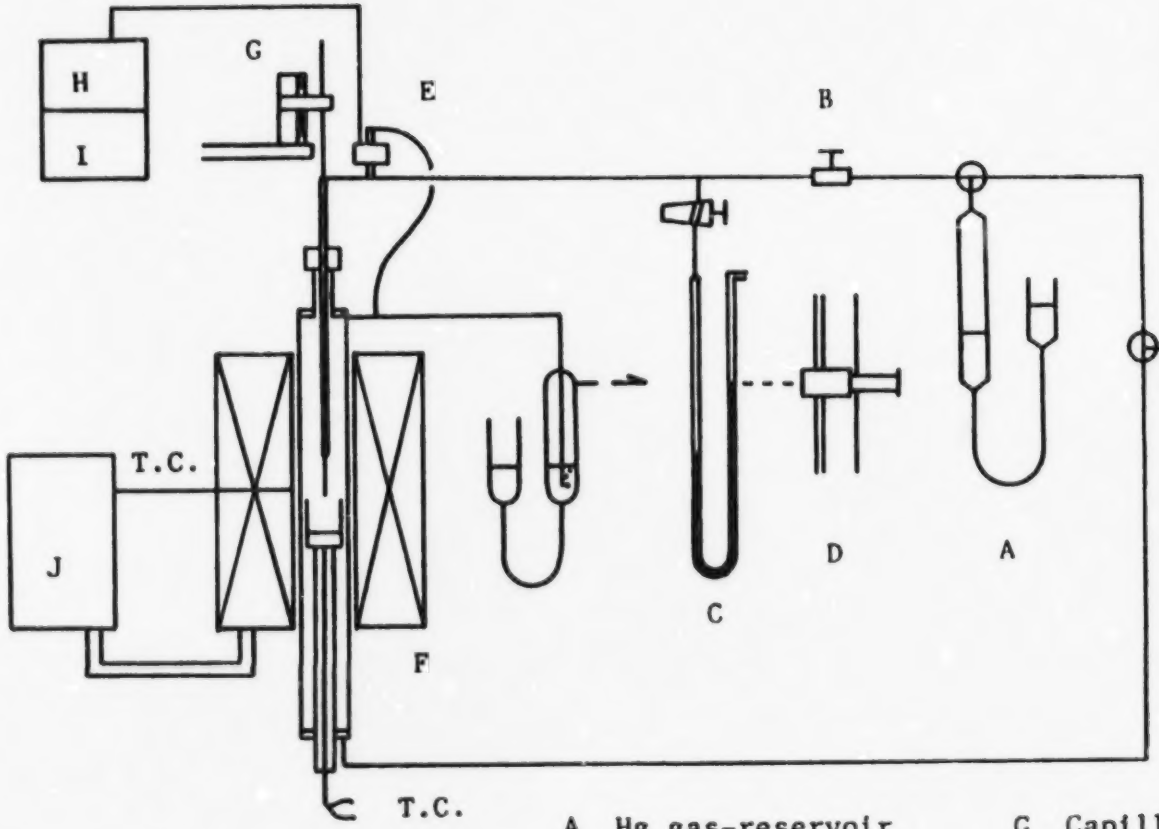
γ is the surface tension, r is the radius of the capillary, P_γ is

the maximum pressure difference inside and outside the bubble at the level of the end of the tip, ρ is the density difference between the liquid salt and the gas and g is the acceleration of gravity.

Apparatus

The overall apparatus used in this investigation is shown in Fig. 1. The furnace had a uniform temperature(2.5 ^{o}C) region of 5 cm. The salt mixtures were contained in graphite crucibles with an inner diameter of 26 mm and a height of 47 mm under a purified argon atmosphere(Fig. 2). The capillary quartz tubes were 5 cm long, with an outer diameter of about 2.1 mm and with an inner diameter of about 1.03 mm. In order to obtain precise results, it was necessary to pay careful attention to the capillary tip. The bore must be as round as possible, the end flat and perpendicular to the axis of the capillary, and the edge of the orifice very sharp. A microscope was used to check the condition and the diameter of the orifice. All measurements were corrected for thermal expansion of the capillary orifice.

As shown in Fig. 3, the quartz capillary tube was connected to an alumina tube with alumina cement(Ceramabond #503). This assembly was connected to a stainless steel tube which could be moved slowly (0.417 mm/min) up or down with a motor(see Fig. 2). The immersion depth of the capillary into the liquid was measured with a cathetometer and also calculated from the rotating speed of the motor. The pressure inside the capillary tube was measured with a pressure transducer (Model PL283TC-2.5-350, Stathan



A Hg gas-reservoir
B Needle valve
C Water manometer
D Cathetometer
E Pressure transducer
F SiC resistance furnace
G Capillary driving device
H Digital volt-meter
I Recorder
J Temperature controller

Figure 1. Surface Tension Apparatus

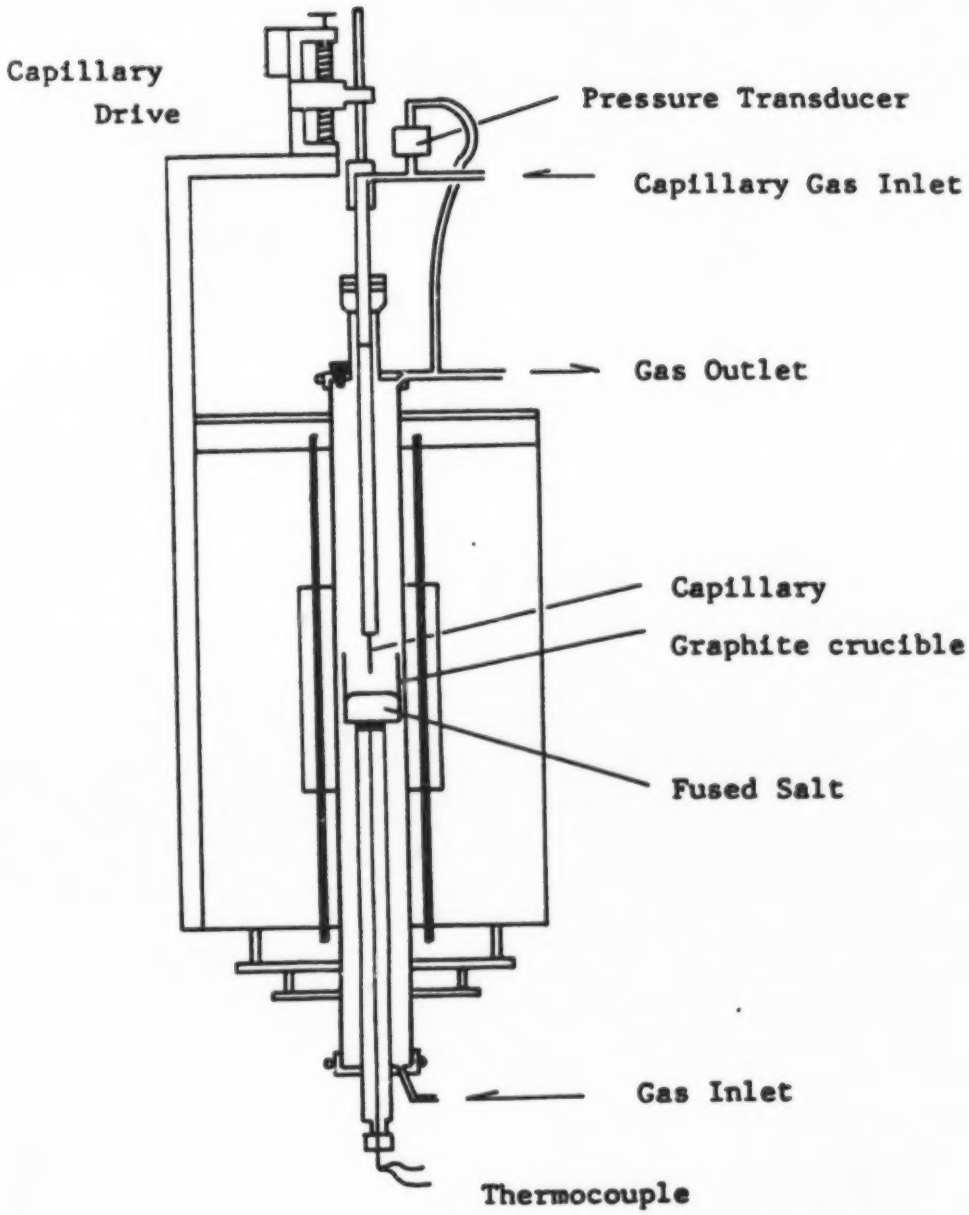


Figure 2. Furnace Assembly

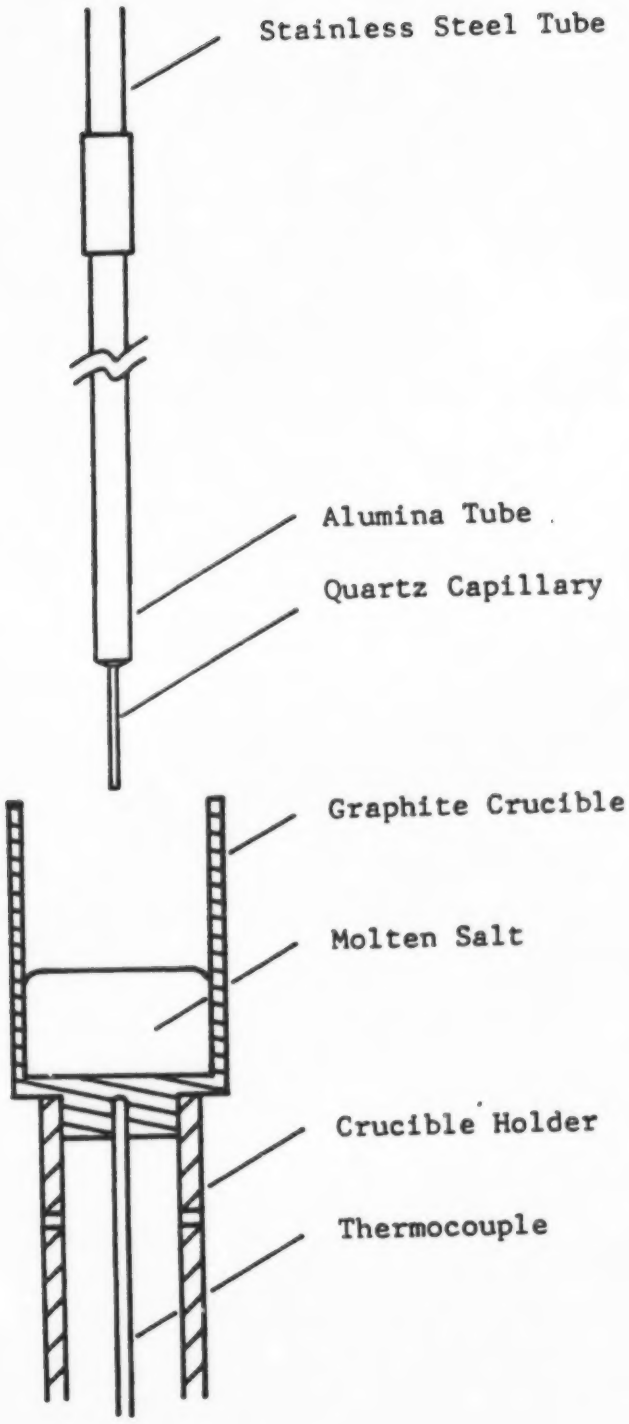


Figure 3. Capillary Assembly

Instruments Inc., Los Angeles) with a sensitivity of 0.001 mV corresponding to 1 Pa. The high purity argon(99.999% min.) used to form the bubbles was passed through a copper getter furnace and through two drying columns packed with drierite and P_2O_5 to remove any possible traces of oxygen and water.

Materials

The potassium chloride(Fisher A.C.S. grade) and the sodium chloride(Fisher U.S.P. grade) were obtained from Fisher Scientific Corporation. The lead chloride was "Baker Analysed" reagent material obtained from J.T. Baker Chemical Co. Each salt was pre-dried in a vacuum oven at 150 ^{o}C. As required, certain amounts of each salt were mixed together to form the desired salt mixture. This mixture was placed into a graphite crucible located in the purification furnace and kept at about 180 ^{o}C under vacuum(1 Pa) for about 10 hrs. Argon was then introduced and the salt mixture heated to just above the melting point. Chlorine gas was bubbled through the melt for about 1 hr. to remove any oxides. This was followed by bubbling of purified argon gas through the melt for about 1 hr. to remove any dissolved chlorine. The melt was then quenched and stored in a desiccator until required. The vaporization loss of lead chloride during the purification treatment was found to be negligibly small.

Procedure

A graphite crucible with the premixed, purified salt mixture was positioned inside the furnace. The capillary tip was cleaned with alcohol and dried with an argon gas jet before it was placed

about 5 cm above the surface of the salt mixture. Each capillary tip was used for only one experiment. The furnace was then flushed with argon and slowly heated up to the desired temperature. Excess pressure was applied to the capillary while it was lowered towards the melt surface. The capillary was then lowered very slowly until a sudden pressure jump was registered by the pressure transducer as shown in Fig. 4, indicating that the capillary had touched the melt surface. The capillary was then stopped and the surface position was recorded. The bubbling rate was adjusted to about 1 bubble/min. which was in the region where the transducer output was independent of the gas flowrate as shown in Fig. 5. The pressure was increased linearly with the immersion depth until a bubble was released at the capillary tip followed by a sudden decrease in the pressure. The capillary immersion depth was changed several times while the maximum bubble pressure was recorded. The pressure drop caused by the formation of a bubble at the capillary tip is the difference in the pressure just before the bubble bursts and after it has burst as shown by the sawtoothed curve in Fig. 4. By measuring the maximum bubble pressure at different immersion depths, the density of the salt can be calculated from the slope of the curve formed when the maximum pressure is plotted as a function of immersion depth as shown in Fig. 6. From the same plot, the pressure drop caused by the formation of the bubble can be found as the maximum pressure for zero immersion depth. Using this pressure value the surface tension of the salt mixture can be calculated using Schrodinger's equation. The temperature effect on the surface tension and the density were determined during both heating and cooling cycles.

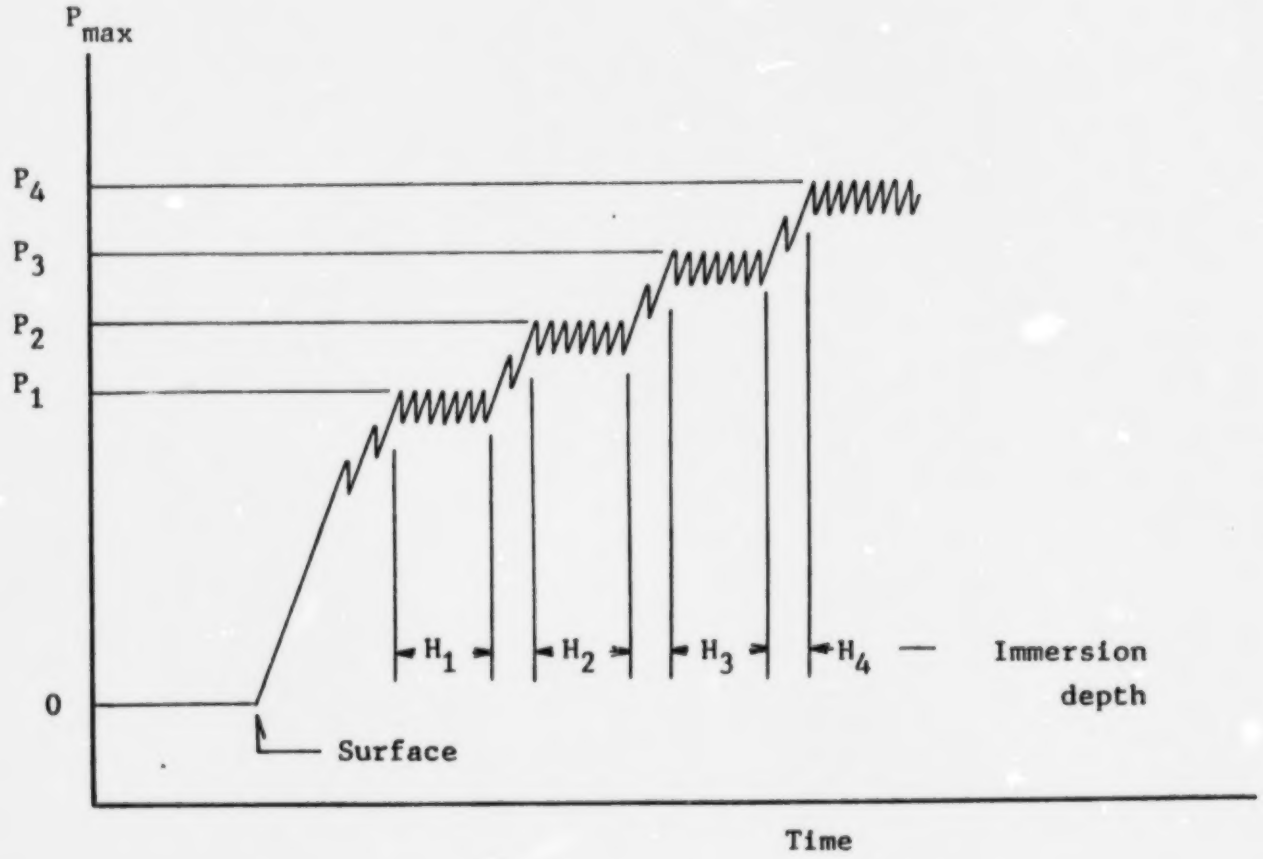


Figure 4. Typical Pressure Transducer Output

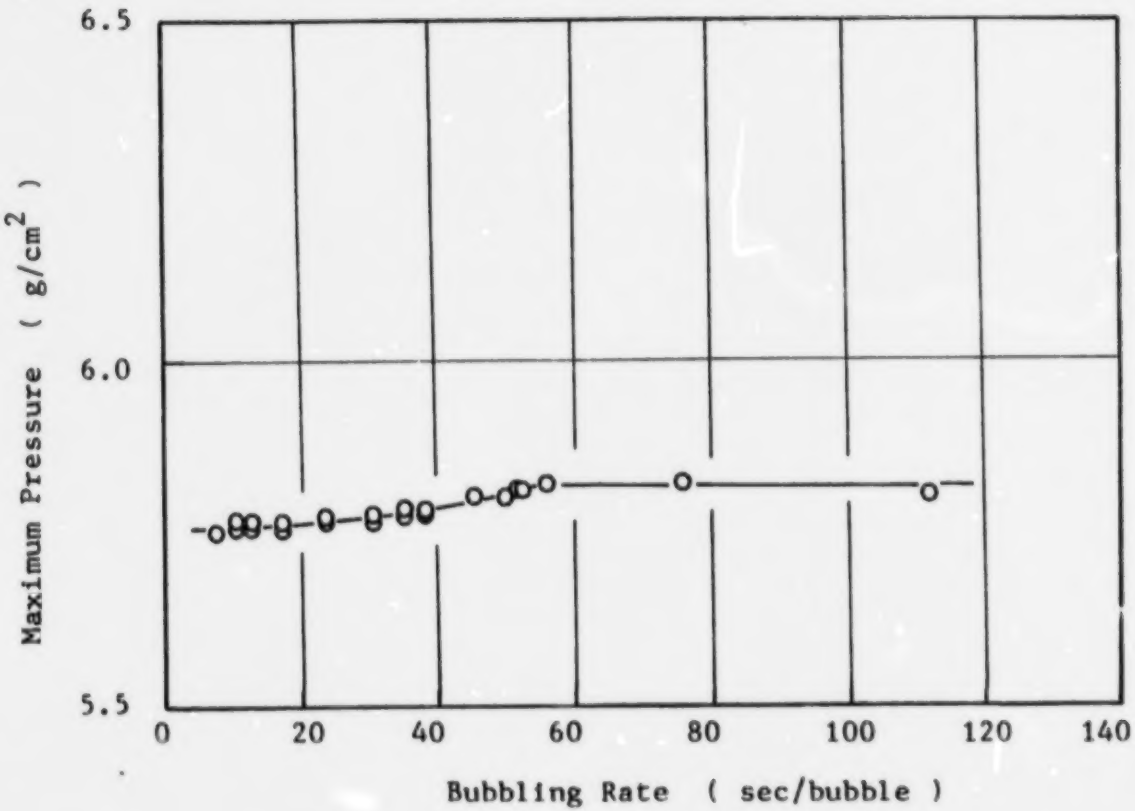
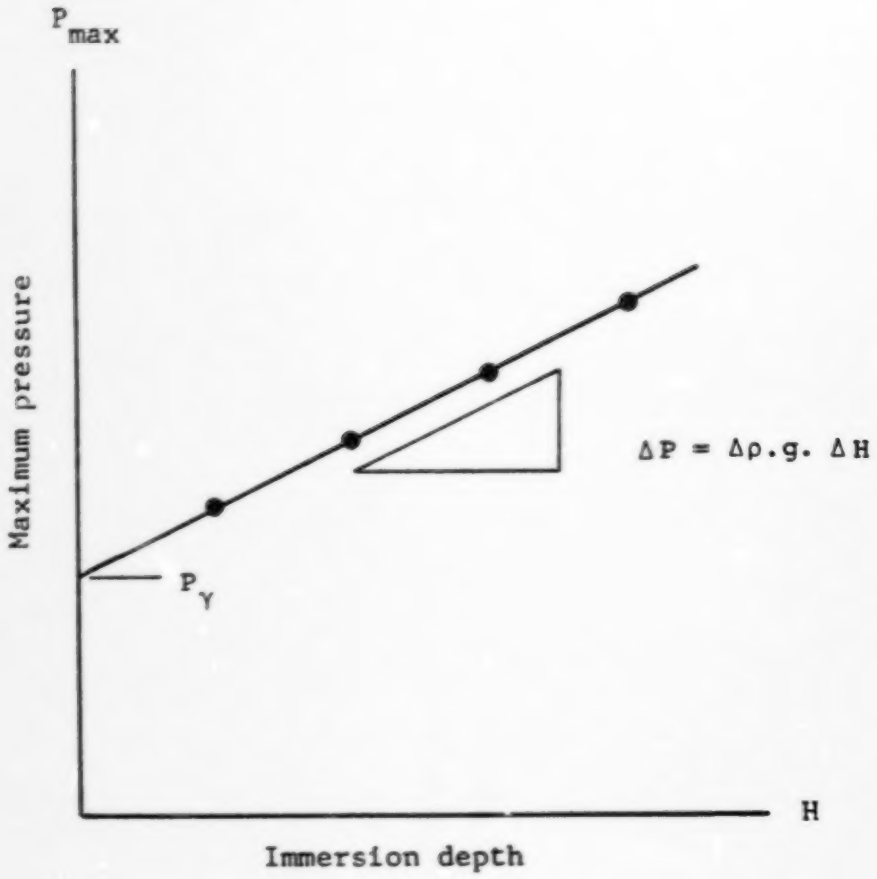


Figure 5. Effect of bubble rate on maximum pressure



Schrödinger's equation

$$\gamma = \frac{r.P_\gamma}{2} \left\{1 - \frac{2}{3}\left(\frac{r.\rho.g}{P_\gamma}\right) - \frac{1}{6}\left(\frac{r.\rho.g}{P_\gamma}\right)^2\right\}$$

Figure 6. Effect of immersion depth on maximum pressure

A typical linear relationship between the maximum pressure and immersion depth is shown in Fig. 7 for a melt containing 60 mole percent $PbCl_2$ and 40 mole percent KCl. The effect of increasing the temperature is to decrease the maximum bubble pressure for a constant depth of immersion.

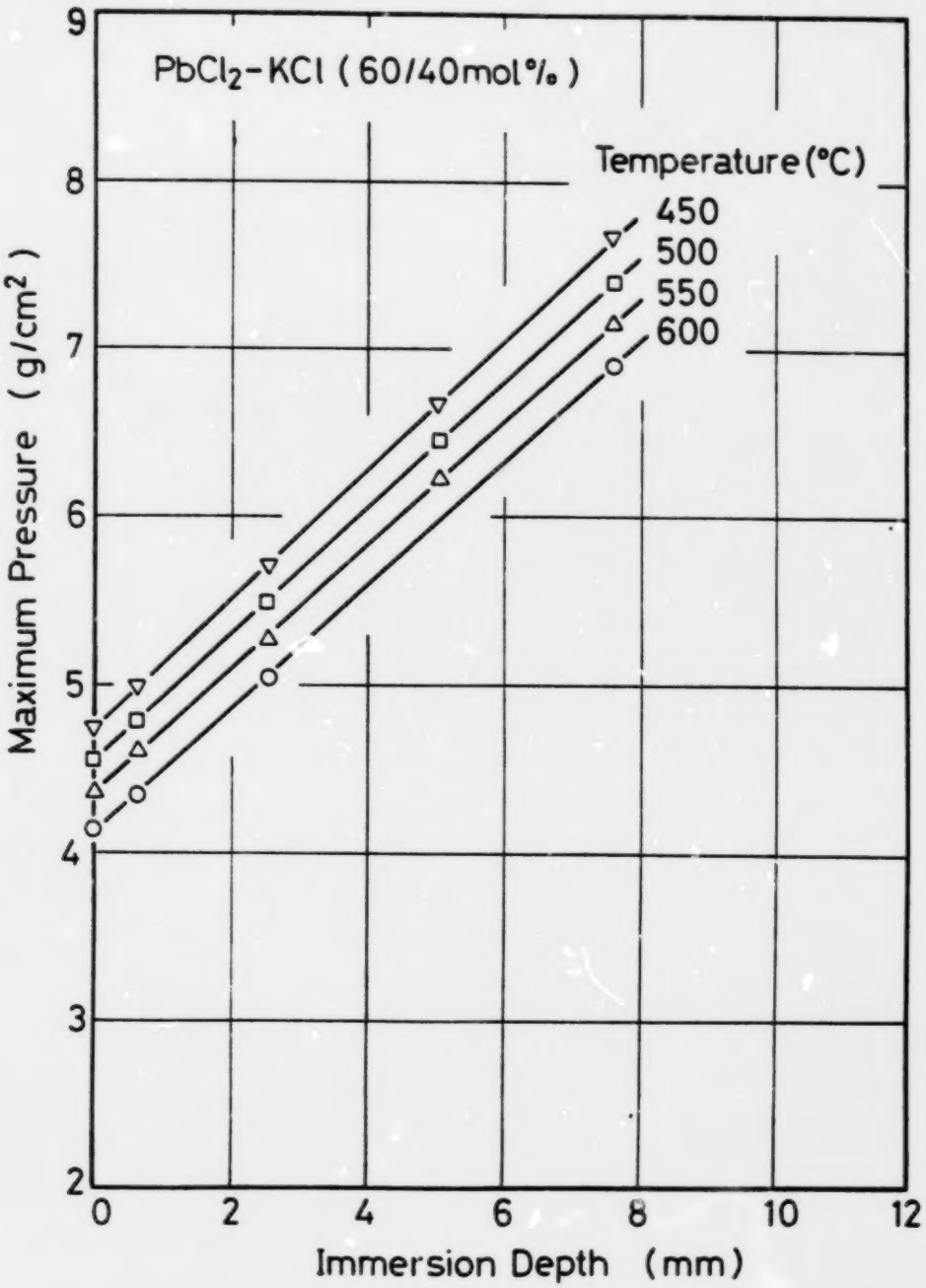


Figure 7. Effect of immersion depth on the maximum pressure- $PbCl_2$-KCl system

Results

The maximum bubble pressure apparatus was tested by measuring the surface tension and density of a 50 mole % mixture of NaCl - KCl. The results of this study are shown in Fig. 8. The surface tension obtained with the present apparatus is 2.5 % lower than the data of Desyatnikov(7) which are recommended by Janz et al(9). Desyatnikov also used the maximum bubble pressure method. On the other hand, the density data obtained are in excellent agreement with both the recent data of Gutierrez and Toguri(4) and the older data of Van Artsdalen and Yaffe(10).

Based on these results, the surface tension and the density of pure $PbCl_2$ and the binary and ternary mixtures of $PbCl_2$, KCl and NaCl have been measured within the temperature range of 450 to 600 ^{o}C.

In the case of pure $PbCl_2$, the experiments were performed at two temperatures, 550 and 600 ^{o}C. The upper limit was dictated by the vapourization loss of $PbCl_2$ and the lower limit by its melting point. Within the experimental temperature range the surface tension can be represented by the following equation:

$$\gamma = 227.9 - 0.122T \text{ (mN/m)}$$

As shown in Fig. 8, the present results are approximately 3.7 %

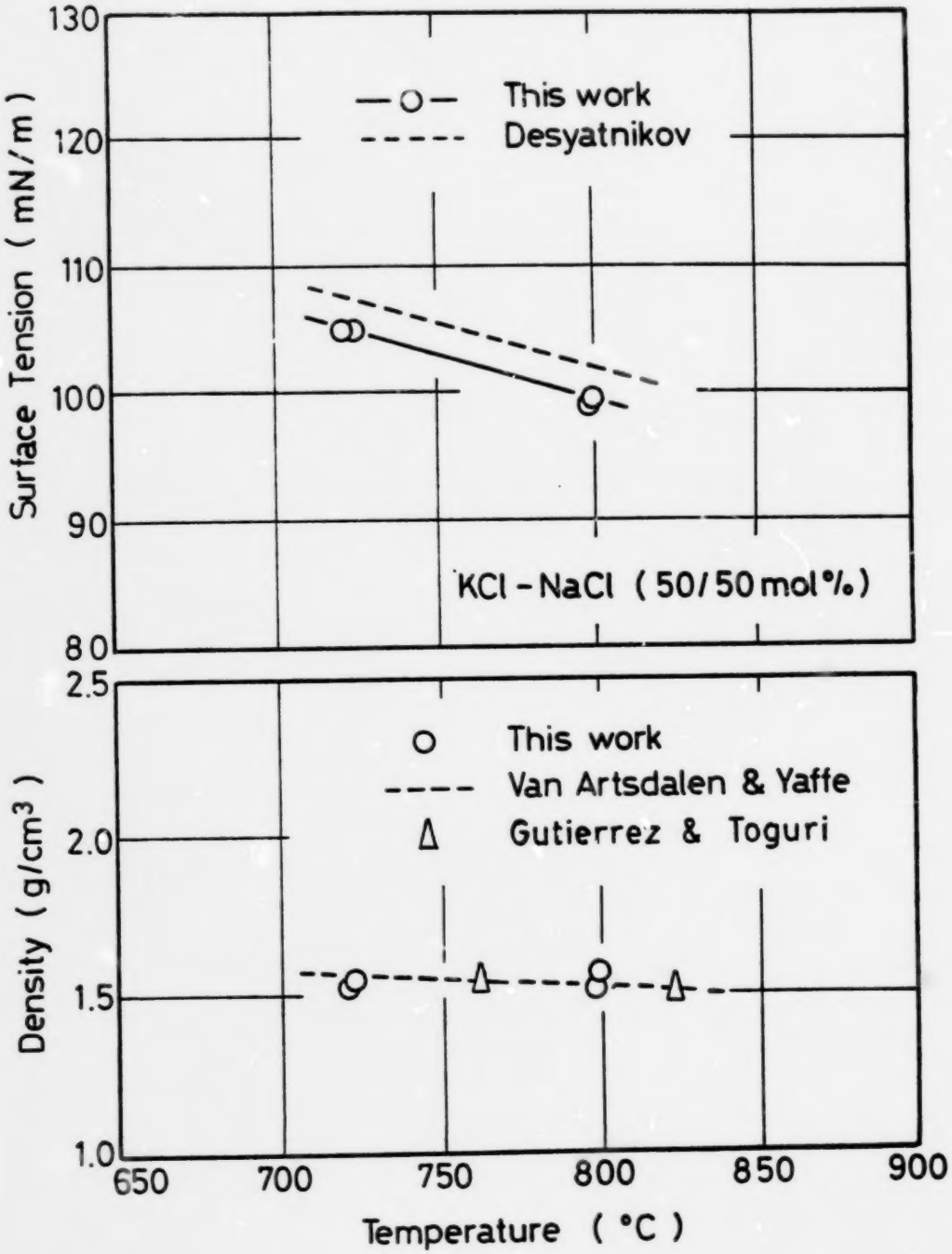


Figure 8. Surface tension and density of KCl-NaCl (50/50 mole ratio) as a function of temperature

below those reported by Dahl and Duke(11) which are shown by the dotted line and which were obtained using the maximum bubble pressure method.

The effect of temperature on the surface tension of melts containing equimolar mixtures of KCl and NaCl with varying amounts of $PbCl_2$ is also shown in Fig. 9. The results indicate that the surface tension decreases linearly with increasing temperature and decreases with decreasing amount of $PbCl_2$ for melts containing more than 40 mole % $PbCl_2$. The corresponding density values are given in Fig. 10. The density for pure $PbCl_2$ obtained in the present study is in good agreement with that of Boardman et al(12) . The effect of temperature on the density of ternary melts is also illustrated and shows a linear decrease with increasing temperatures. Good agreement is found with the reported values of Gutierrez and Toguri(4) as shown by the dotted lines.

The effect of the concentration of $PbCl_2$ on the surface tension for an equimolar KCl-NaCl melt is shown in Fig. 11. A decrease in the $PbCl_2$ concentration lowers the surface tension. The corresponding density values are shown in Figs. 12 and 13 for 550 and 600 ^{o}C respectively. The density decreases with decreasing $PbCl_2$.

The surface tension and the density were also measured for mixtures with a constant amount of $PbCl_2$(60 mole %) and with varying amounts of KCl and NaCl. Figure 14 shows the effect of NaCl addition on the surface tension at various temperatures. The effect of temperature is indicated in Fig. 15. The corresponding

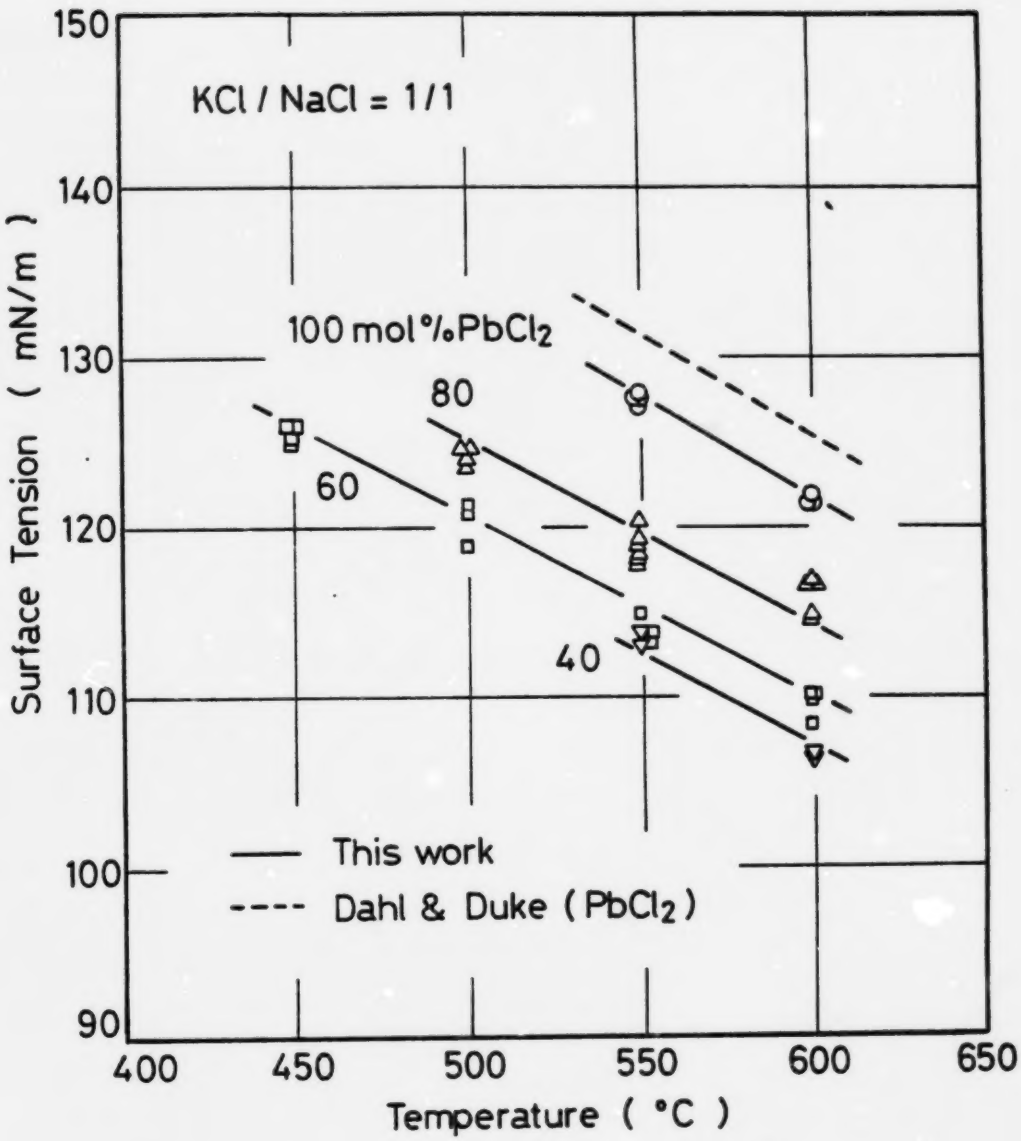


Figure 9. Temperature dependency of surface at constant KCl/NaCl molar ratio in the $PbCl_2$-KCl-NaCl system

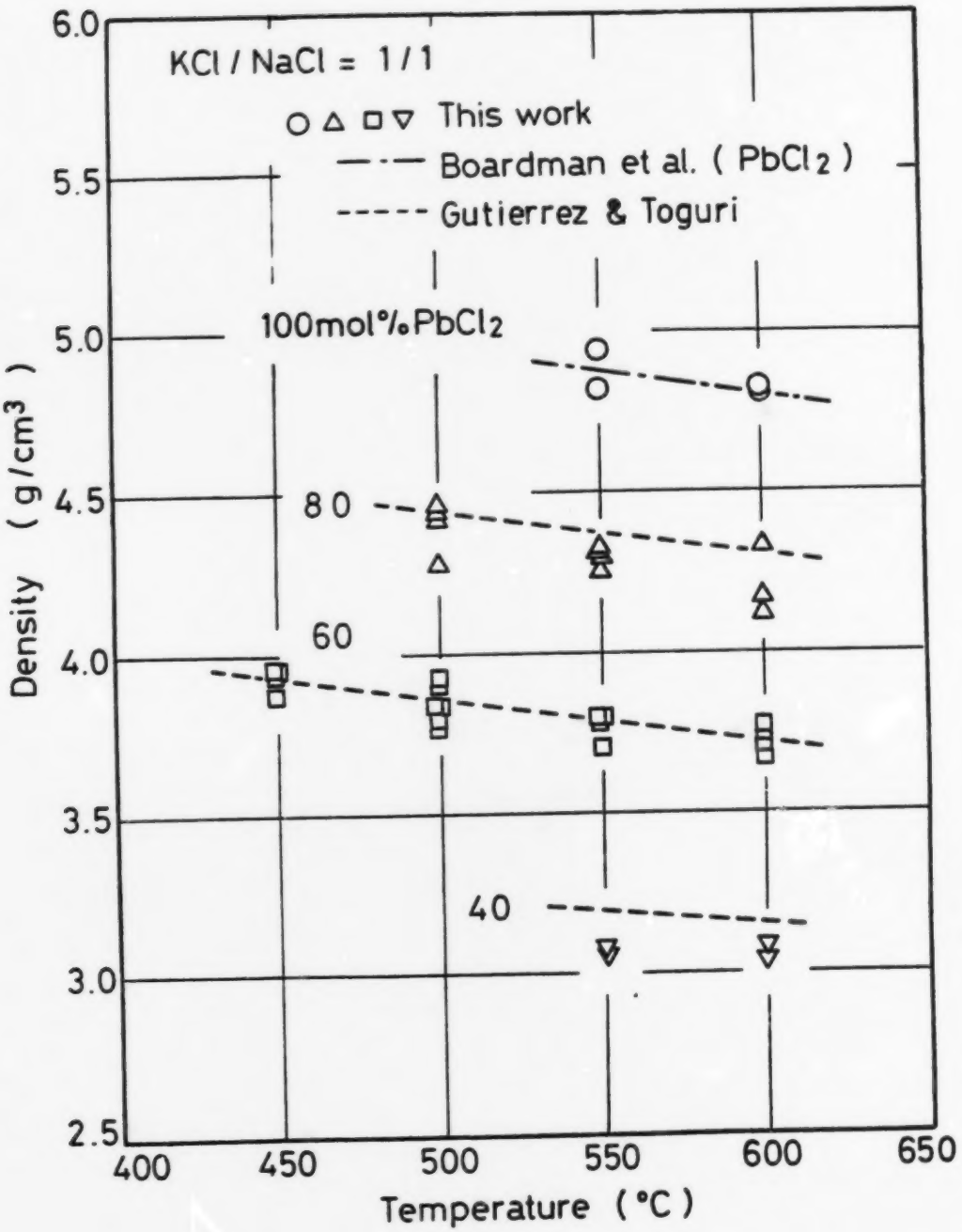


Figure 10. Temperature dependency of density at constant KCl/NaCl molar ratio in the $PbCl_2$-KCl-NaCl system

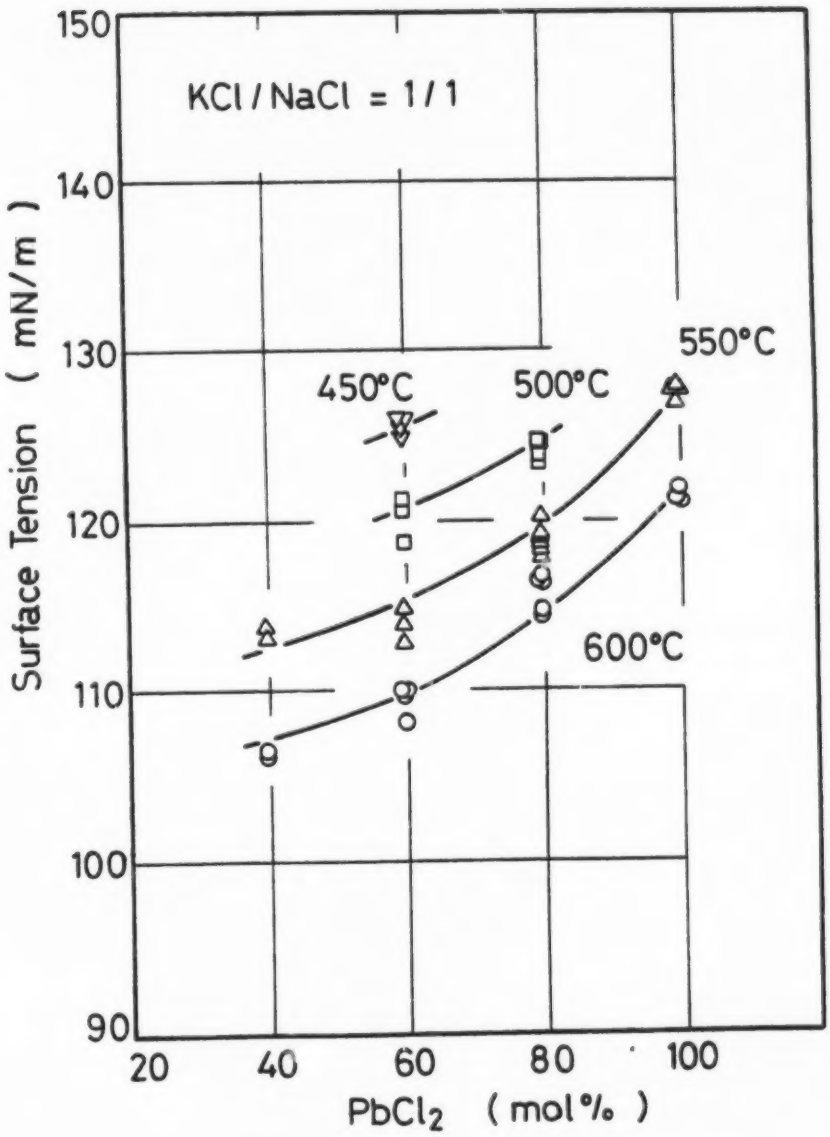


Figure 11. Composition dependency of surface tension at constant KCl/NaCl mole ratio in the $PbCl_2$-KCl-NaCl ternary system

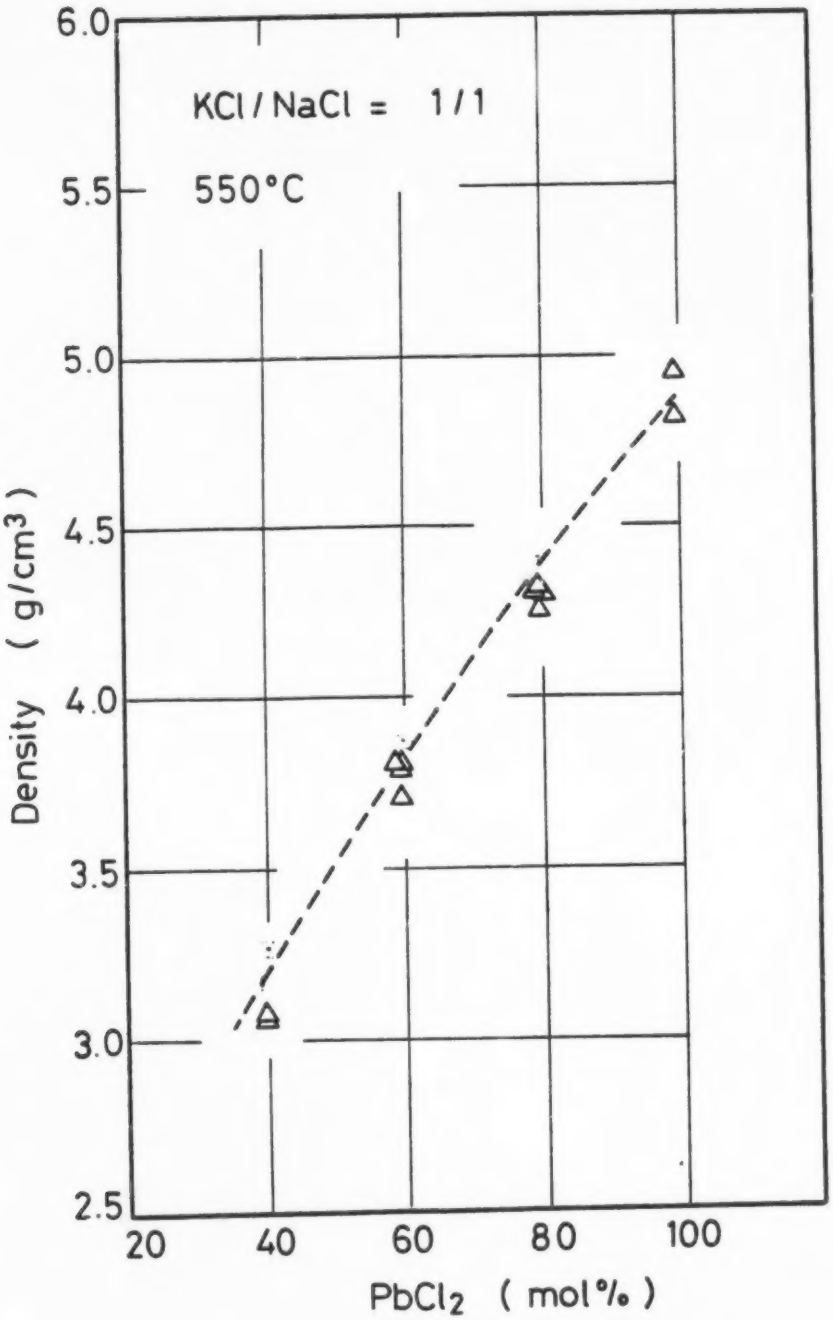


Figure 12. Composition dependency of density at constant KCl/NaCl molar ratio in the $PbCl_2$-NaCl-KCl ternary system

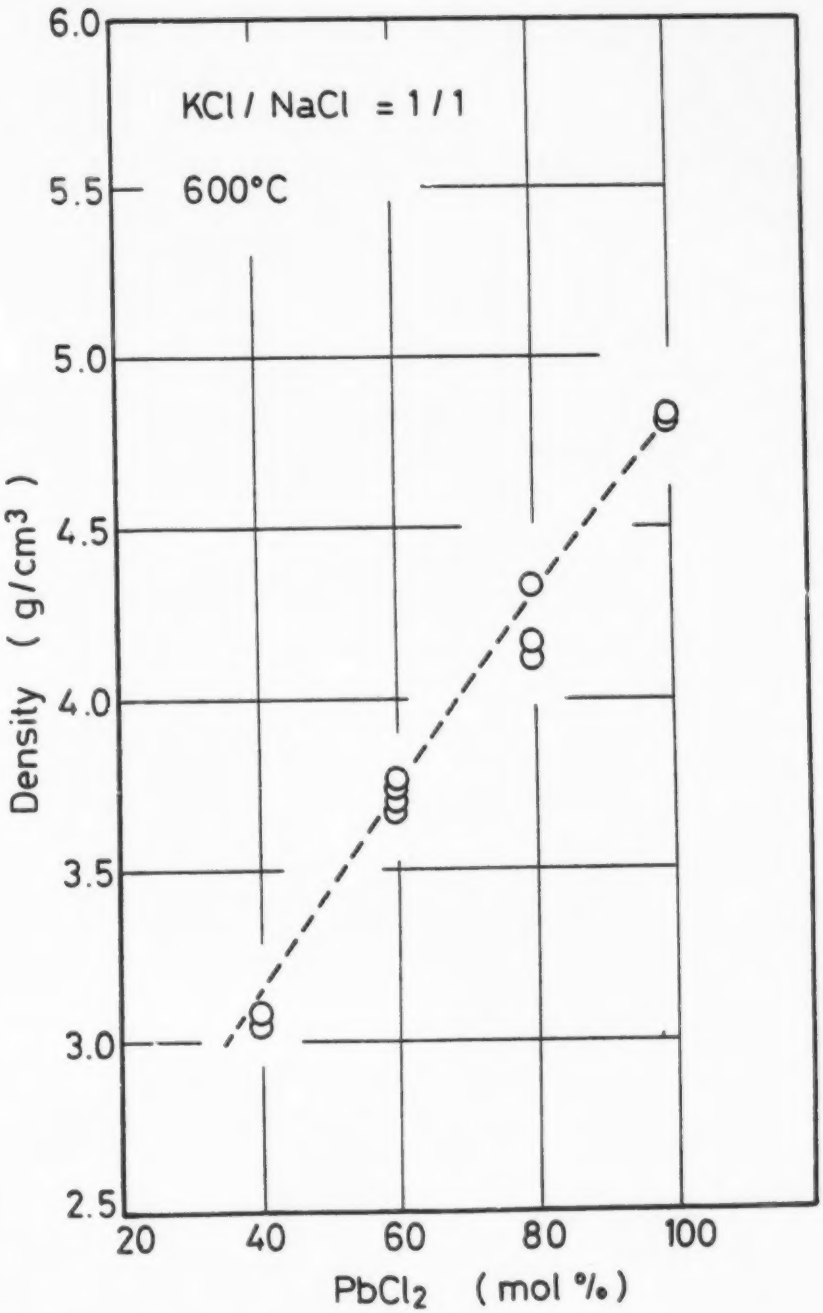


Figure 13. Composition dependency of density at constant KCl/NaCl molar ratio in the $PbCl_2$-KCl-NaCl ternary system

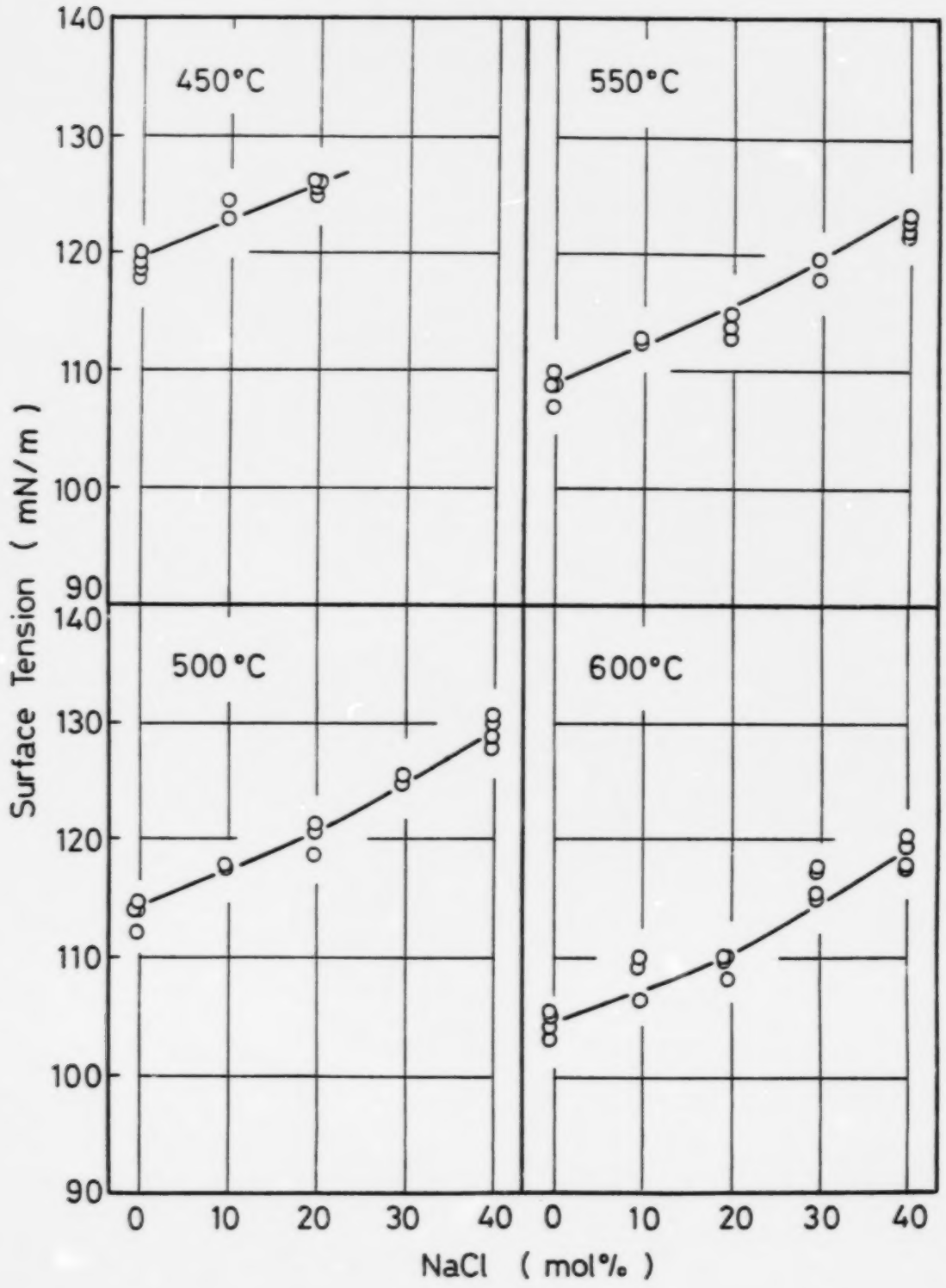


Figure 14. Composition dependency of surface tension at constant $PbCl_2$ content(60 mole percent) in the $PbCl_2$-KCl-NaCl ternary system

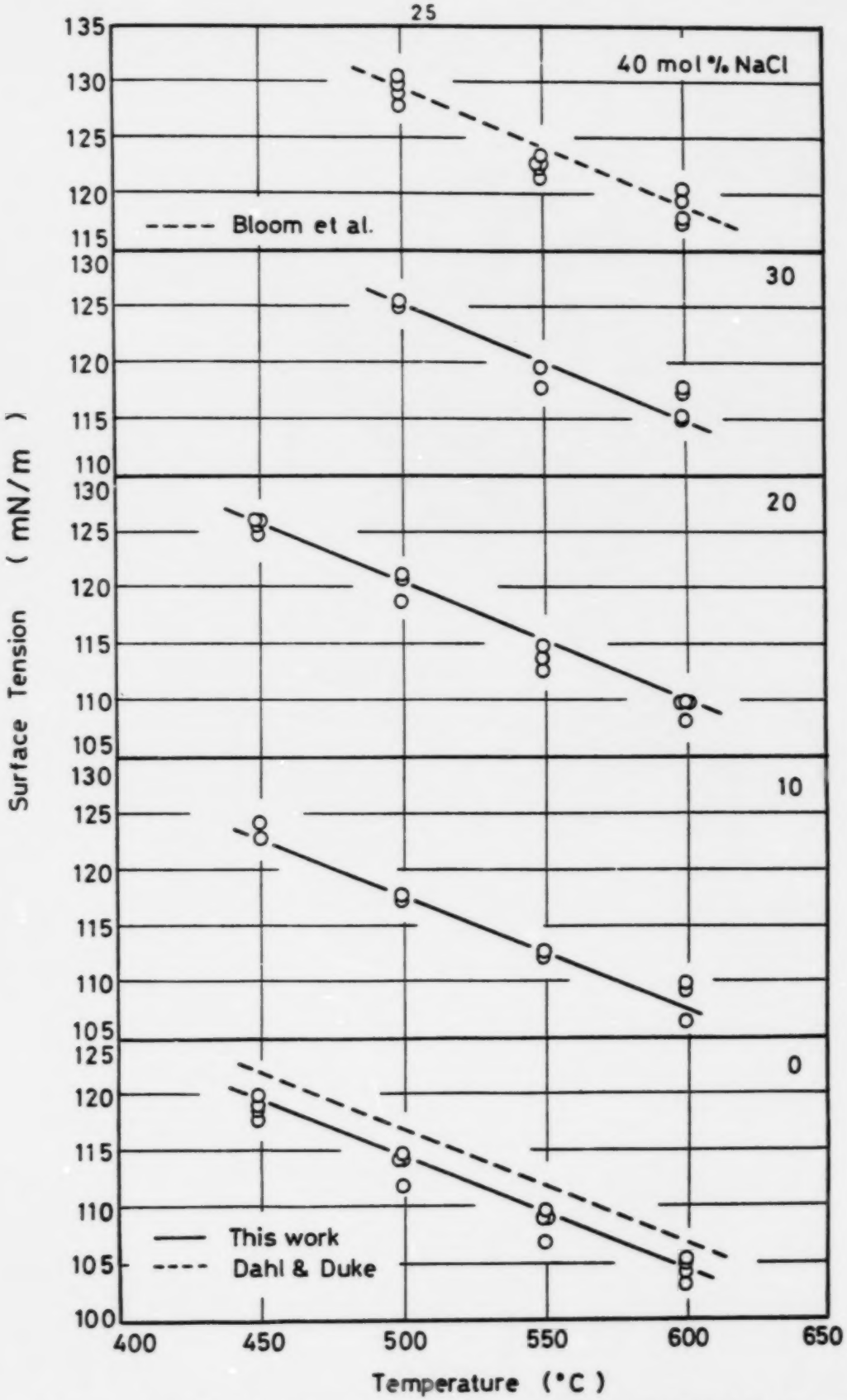


Figure 15. Temperature dependency of surface tension at constant $PbCl_2$ content (60 mole percent) in the $PbCl_2$-KCl-NaCl ternary system

densities are illustrated in Fig. 16.

The surface tension of the binary mixture of $PbCl_2$-NaCl is higher than that of a mixture containing $PbCl_2$ and KCl. From the obtained data, an almost linear relation can be observed between the surface tension of the ternary mixtures and the ratio of KCl to NaCl. Similar relations are observed also for the density values with the highest density values in the $PbCl_2$-NaCl binary.

The compositions of the ternary system can be expressed by the parameters y and t , where

$$y = x_{PbCl_2}$$

$$t = x_{KCl}/(x_{KCl} + x_{NaCl})$$

The average surface tension values at each temperature and composition are given in Table 1 together with the fitted coefficients of the linear equation

$$\gamma = A + B*T(K) \quad (mN/m)$$

The corresponding average density values are given in Table 2 together with the fitted coefficients of the linear equation

$$\rho = C + D*T(K) \quad (g/cm^3)$$

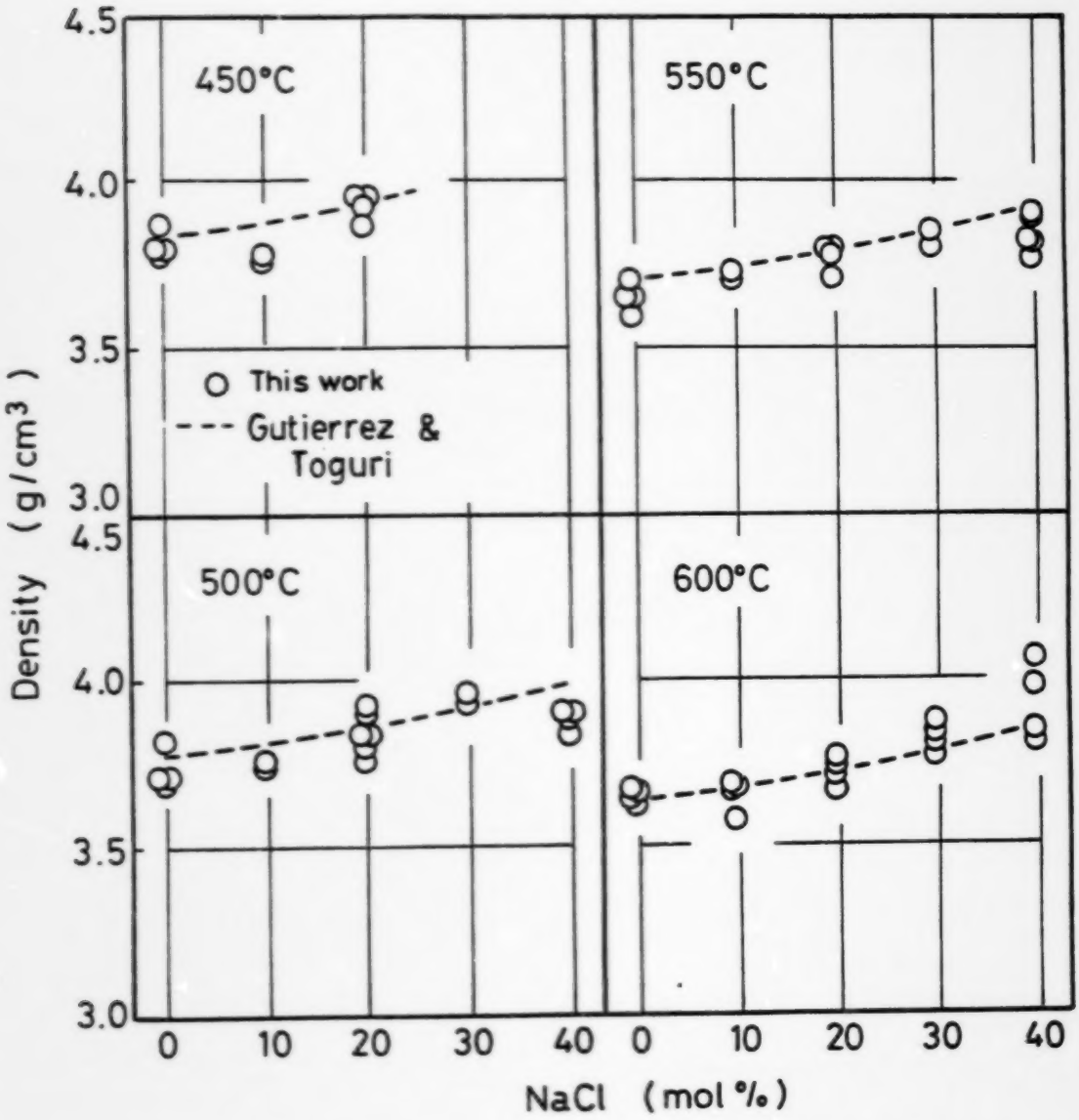


Figure 16. Composition dependency of density at constant $PbCl_2$ content(60 mole percent) in the $PbCl_2$-KCl-NaCl ternary system

Table 1. Surface tension for the $PbCl_2$-KCl-NaCl system

Composition		Temperature				$\gamma = A + B*T(K)$	
y	t	723K	773K	823K	873K	A	B
1.0		-	-	127.5	121.4	227.9	-0.122
0.8	0.5	-	124.1	118.8	115.8	187.9	-0.083
0.6	1.0	119.4	114.4	109.4	103.7	194.9	-0.104
0.6	1.0	118.5	113.0	107.9	105.2	183.0	-0.090
0.6	0.75	123.7	117.7	112.5	108.6	196.2	-0.101
0.6	0.5	125.2	-	-	109.2	202.3	-0.107
0.6	0.5	126.1	120.3	113.4	109.9	206.0	-0.111
0.6	0.25	-	125.2	118.8	116.4	192.6	-0.088
0.6	0.0	-	130.3	123.4	-	237.0	-0.138
0.6	0.0	-	128.4	122.2	118.9	201.4	-0.095
0.4	0.5	-	-	113.5	106.5	228.7	-0.140
0.0	0.5	105.1(996 K)		99.3(1073 K)		179.8	-0.075

y: mole fraction of $PbCl_2$

t: mole fraction of KCl over sum of mole fractions of KCl and NaCl

Surface tension(γ) is given in mN/m

Table 2. Density for the $PbCl_2$-KCl-NaCl ternary system

Composition		Temperature				$\rho = A + B*T(K)$	
y	t	723K	773K	823K	873K	A	10^4B
1.0		-	-	4.89	4.82	6.04	-14.0
0.8	0.5	-	4.40	4.29	4.21	5.86	-14.0
0.6	1.0	3.80	3.71	3.65	3.63	4.58	-11.0
0.6	1.0	3.82	3.75	3.65	3.67	4.60	-11.0
0.6	0.75	3.77	3.75	3.65	3.67	4.28	-7.0
0.6	0.5	3.94	-	-	3.71	5.02	-15.0
0.6	0.5	3.92	3.80	3.75	3.74	4.76	-12.0
0.6	0.25	-	3.95	3.83	3.83	4.86	-12.0
0.6	0.0	-	3.91	3.89	-	4.22	-4.0
0.6	0.0	-	3.86	3.85	3.83	4.09	-3.0
0.4	0.5	-	-	3.07	3.06	3.15	-1.0
0.0	0.5	1.54(996 K)		1.55(1073 K)		1.44	+1.0

y: mole fraction of $PbCl_2$

t: mole fraction of KCl over sum of mole fractions of KCl and NaCl

Density is given in g/cm^3

Discussion

Dahl and Duke(11) showed that the surface tension of the lead chloride - alkali chloride binary mixtures have a minimum value at approximately 40 mole percent $PbCl_2$. The extent of this minimum increases as the size of the alkali ion increases(from Na to Cs). A similar minimum in the surface tension was observed in this work. By extrapolating the values of the equimolar binary mixture of KCl and NaCl to 823 and 873 K as shown in Fig. 17, a pronounced minimum can be observed at a $PbCl_2$ content of approximately 40 mole percent. The plotted points are based on the average experimental values and the lines are the least squares fitted second order polynomials. This behaviour corresponds well with that observed by Dahl and Duke(11) and by Sternberg et al(13). This behaviour may be explained by the formation of surface active $PbCl_{2+n}^{n-}$ complexes, where the value of n may be 1,2 or 4. Support for this suggestion comes from studies of Raman spectra(14), surface tension measurements(11)and density measurements(15). Similar complex formation has also been suggested in mixtures of $CdCl_2$ with alkali chlorides based on surface tension measurements(16).

Figure 18 shows the surface tension of ternary mixtures containing a constant amount of $PbCl_2$ (60 mole percent) but with varying amounts of KCl and NaCl at 773 and 873 K. As illustrated by the fitted straight lines, the surface tension of the ternary mixtures can be calculated, within experimental errors from the two binary systems $PbCl_2$ - KCl and $PbCl_2$ - NaCl by the following

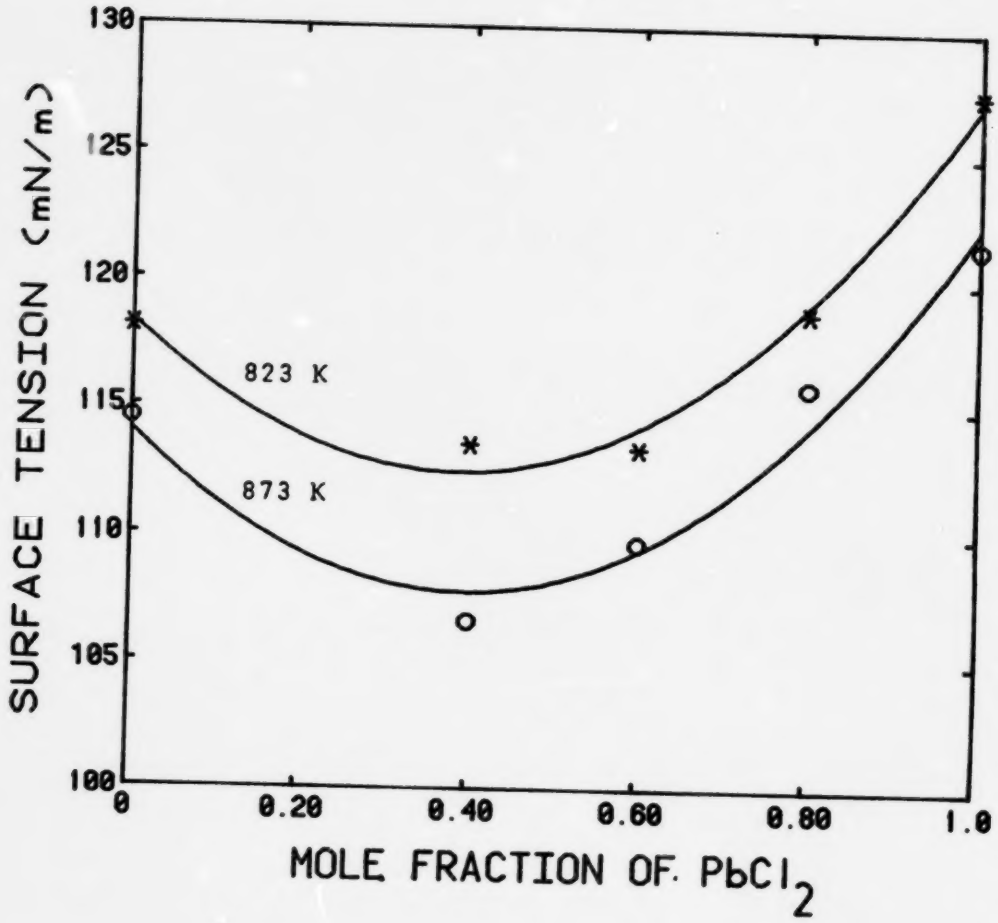


Figure 17. Composition dependency of surface tension at constant KCl/NaCl molar ratio(50/50)

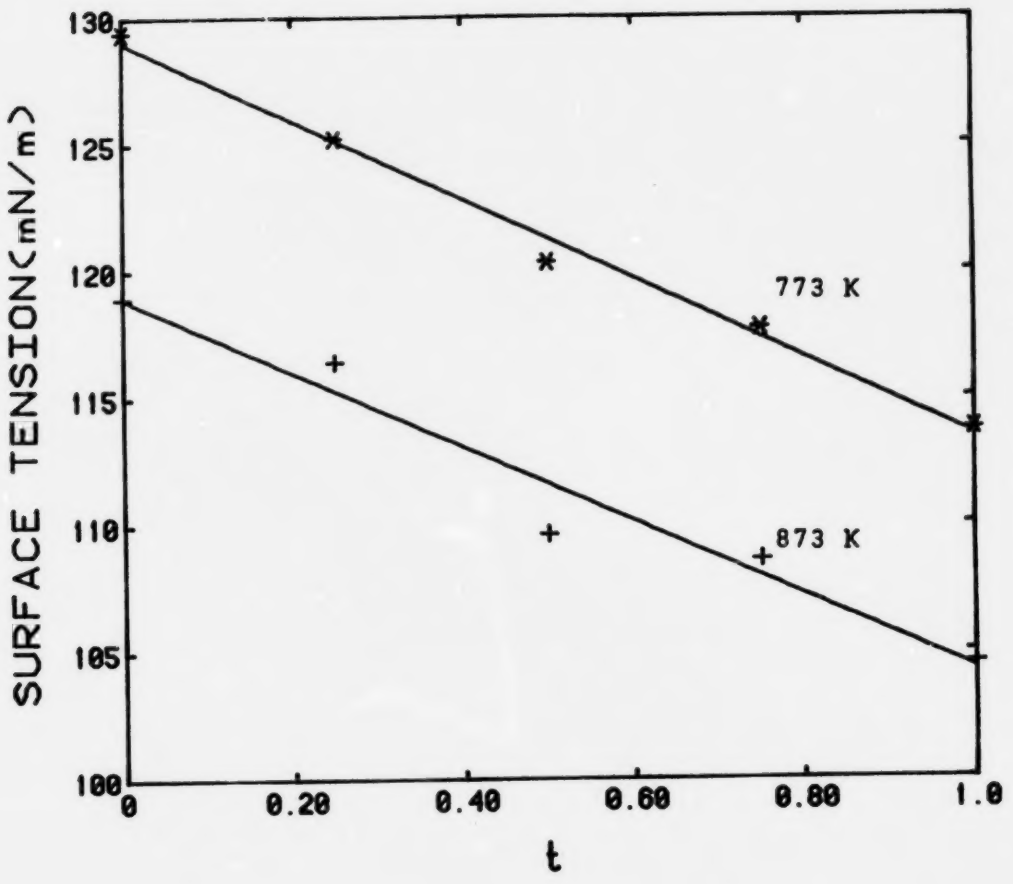


Figure 18. Surface tension of mixtures with 60 mole percent $PbCl_2$ as a function of the KCl/NaCl ratio

$t = {}^{x}KCl/(x_{KCl} + x_{NaCl})$

expression:

$$\gamma(\text{ternary}) = t^*\gamma_{12} + (1-t)^*\gamma_{13}$$

where γ_{12} and γ_{13} are the surface tension of the binaries $PbCl_2$ - KCl and $PbCl_2$ - NaCl, respectively. By assuming that this additive relation is valid throughout the ternary system it is possible to calculate the surface tension of any mixture from the data of the two binaries $PbCl_2$ - KCl and $PbCl_2$ - NaCl. It must however, be noted that the present binary values are approximately 2 % lower than those reported earlier. This additive behaviour has previously been found(4) to be valid for the molar volumes of tenary mixtures of $PbCl_2$, KCl and NaCl. By using reported(9) binary surface tension values, the following equations were found by a least squares curve fitting:

$PbCl_2$ - KCl:

$$\gamma_{12} = 177.35 - 17.82*y + 73.49*y^2 - (6.44 + 5.94*y)*T$$

$PbCl_2$ - NaCl:

$$\gamma_{13} = 218.45 - 39.2*y + 42.02*y^2 - (9.52 + 1.44*y)*T$$

$PbCl_2$ - KCl - NaCl: $\gamma = t^*\gamma_{12} + (1-t)^*\gamma_{13}$

By using these equations, constant surface tension lines can be calculated. Figure 19 shows the liquidus lines and the constant surface tension lines for the $PbCl_2$-KCl-NaCl ternary system at

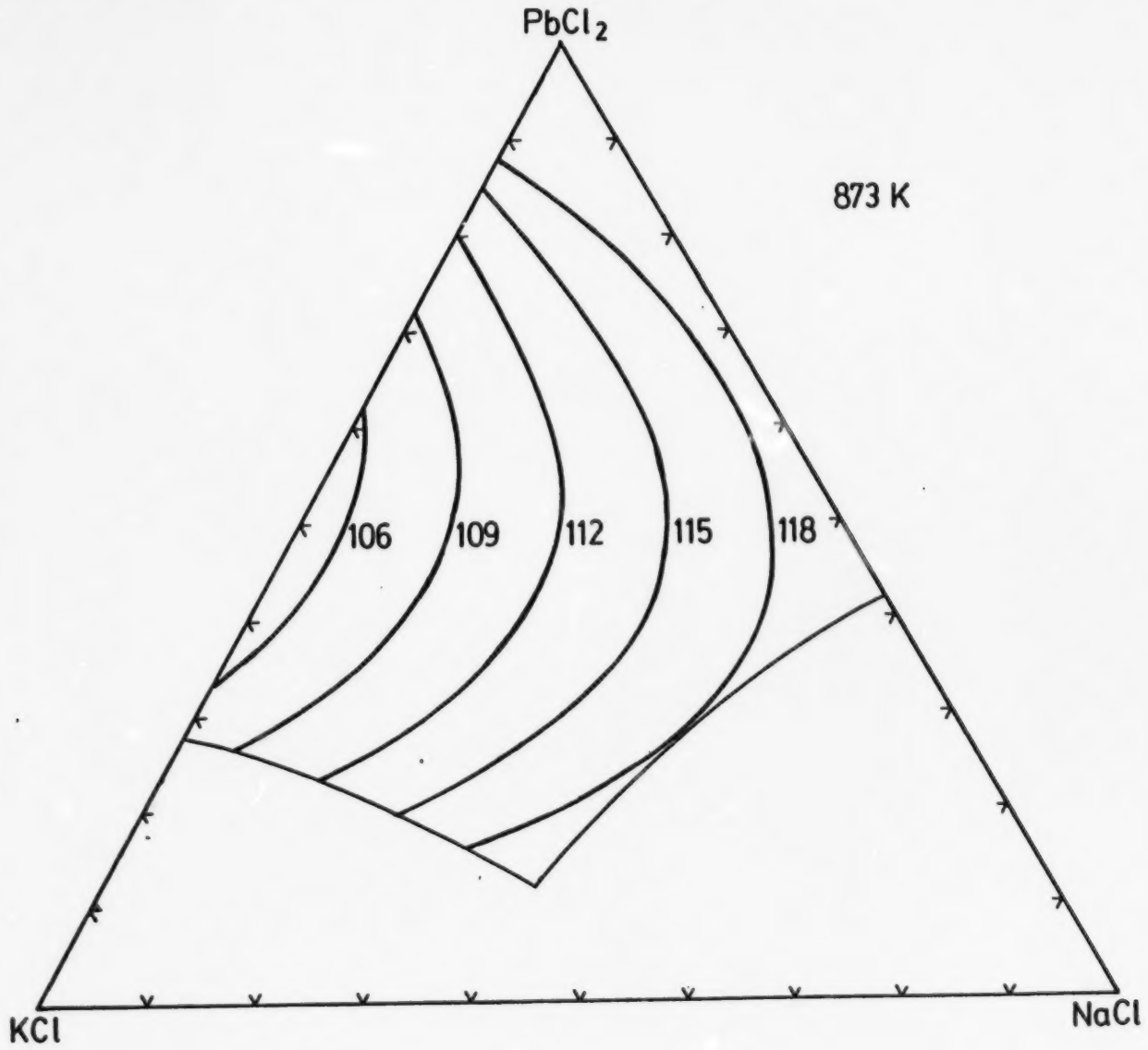


Figure 19. Constant surface tension lines for the $PbCl_2$-KCl-NaCl ternary system at 873 K

873 K. For a constant $PbCl_2$ content the surface tension increases on going from the KCl to the NaCl rich side. For a given KCl to NaCl ratio a minimum in the surface tension is observed at approximately 45 mole % $PbCl_2$, which is slightly higher than that found in this study. The values measured in this work are all within 3 % from those calculated by the above equations with the exception of the mixture of equimolar amounts of KCl and NaCl with 40 mole % $PbCl_2$ where the deviation is 4 %. This is consistent with the present study in which the minimum in the surface tension was obtained for mixtures containing 40 mole % $PbCl_2$. In a similar manner the surface tension of the ternary system $PbCl_2$-KCl-LiCl has been estimated and discussed in Appendix 1.

The density values obtained in this work using the maximum bubble pressure method are not expected to of the same accuracy as those obtained by direct Archimedean methods. Nevertheless, the data obtained in the precent study were in excellent agreement with those of Gutierrez and Toguri(4) who used a bottom-balance Archimedean technique.

CONCLUSIONS

1) The surface tension and densities of the $PbCl_2$-KCl-NaCl ternary system have been determined over the temperature range of 450 to 650 ^{o}C by a maximum bubble pressure method.

2) The surface tension of these melts were found to be additive at constant values of y and this behaviour could be described by the additivity rule with respect to t:

$$\gamma = t^* \gamma_{12} + (1-t)^* \gamma_{13}$$

where the two binary systems, each containing $PbCl_2$, are denoted by the subscripts 12 and 13.

APPENDIX 1. Surface tension of the $PbCl_2$-KCl-LiCl ternary system.

The surface tension of the KCl-LiCl binary system deviates negatively from ideality. The surface tension of the $PbCl_2$-KCl-LiCl ternary system can therefore not be estimated by the simple additive rule. No complex formation is expected to occur in the KCl-LiCl system and the non-ideal behaviour of the surface tension is caused by surface enrichment of the species with the lowest surface tension(KCl).

Literature(5,17) surface tension values of the KCl-LiCl binary at 980 K is shown in Table 3 together with those calculated by the following equation:

$$\gamma = \gamma_{KCl} + (\gamma_{LiCl} - \gamma_{KCl})\left(\frac{x_{LiCl}}{x_{KCl} + x_{LiCl}}\right)^2$$

Assuming that a similar relation is valid also for the ternary system, the surface tension can be calculated using the following expression:

$$\gamma = \gamma_{12} + \left(\frac{x_{LiCl}}{x_{KCl} + x_{LiCl}}\right)^2 (\gamma_{13} - \gamma_{12})$$

where γ_{12} and γ_{13} are the surface tensions in the $PbCl_2$-KCl and

$PbCl_2$-LiCl binary systems. The surface tension of the $PbCl_2$-LiCl binary system can be estimated from the following equation:

$$\gamma = 192.88 + 27.64*y + 7.25*y^2 - (7.79 + 3.91*y)*10^{-2}T$$

By using the above equations constant surface tension contours have been calculated for the $PbCl_2$-KCl-LiCl system at 773 K as shown in Fig. 20.

Table 3. Surface tension of the KCl-LiCl binary system at 980 K.

x_{LiCl}	Surface tension (Lit.)	(Calc.)
0.0	103.8	103.8
0.2	105.9	104.5
0.3	106.1	105.3
0.4	106.8	106.5
0.5	108.0	108.1
0.7	111.7	112.2
0.8	114.2	114.7
0.9	117.2	117.7
1.0	120.9	120.9

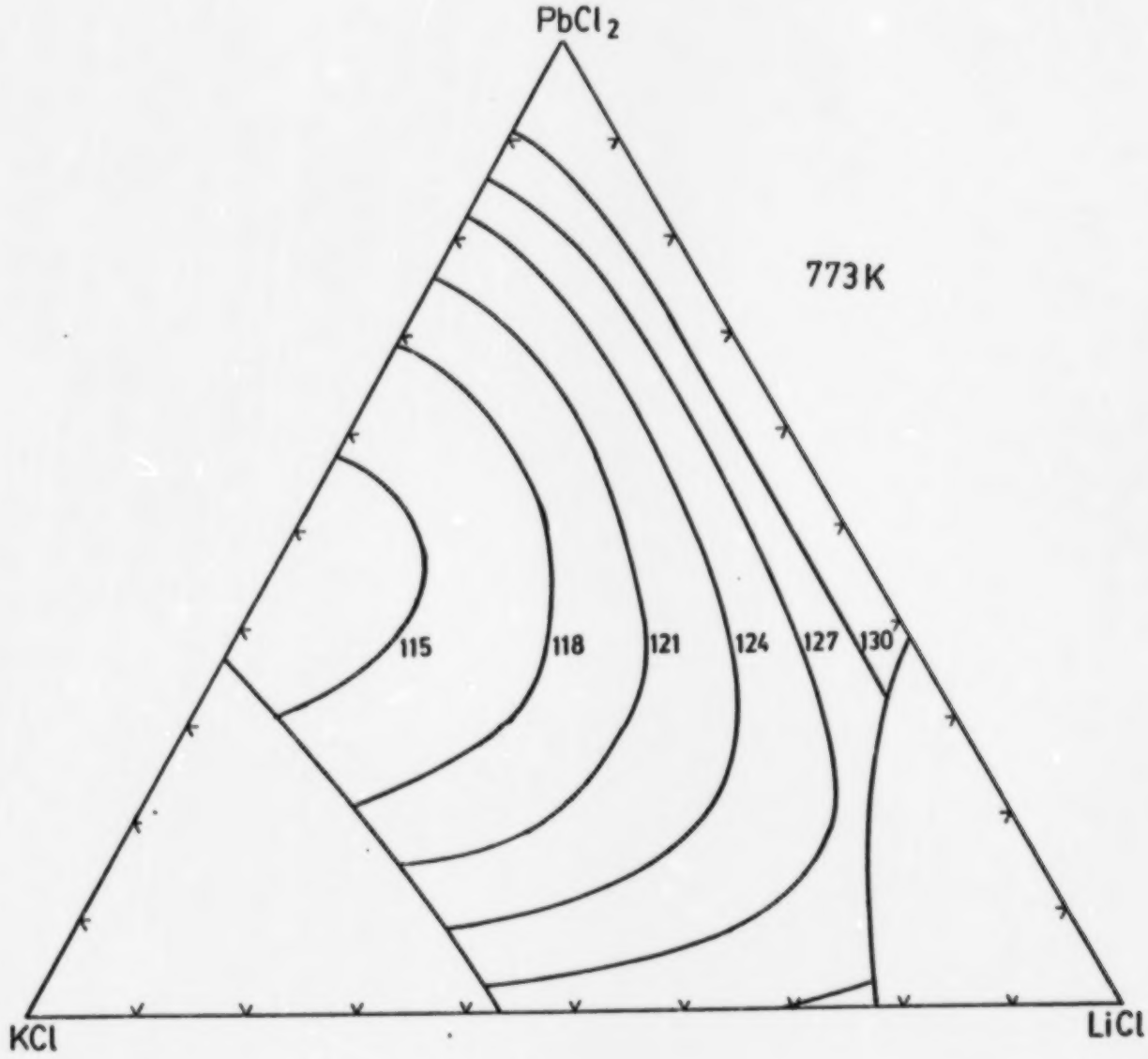


Figure 20. Constant surface tension lines for the $PbCl_2$-KCl-LiCl ternary system at 773 K

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